

## Supporting Information for

### 4-Iodo-1H-imidazole Dramatically Improving the Open-circuit Voltages of Perovskite Solar Cell Exceeding 1.2 V

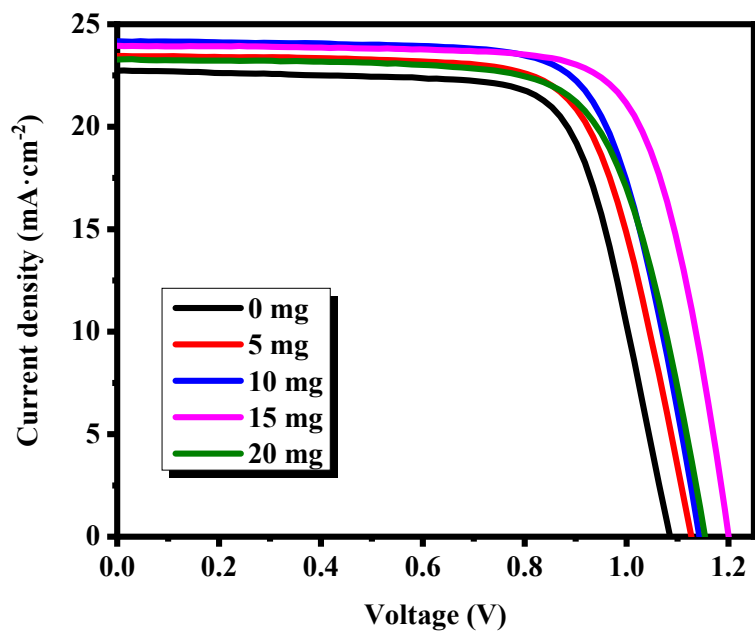
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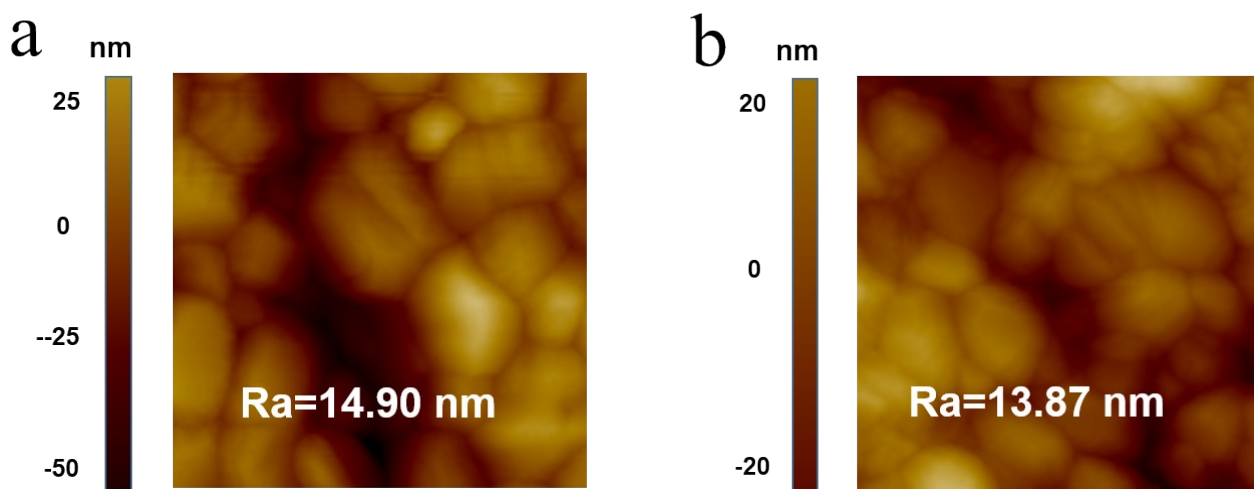
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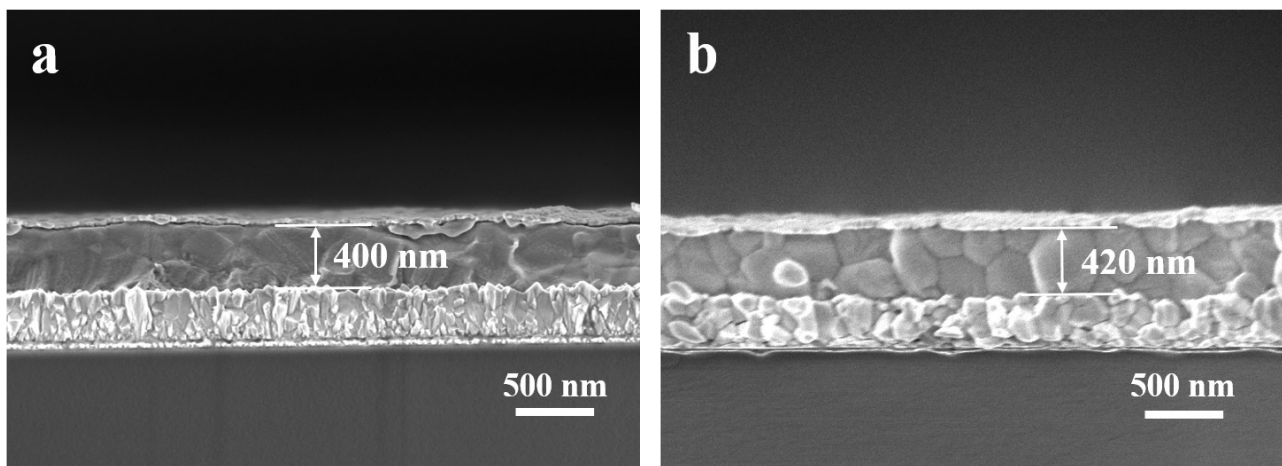
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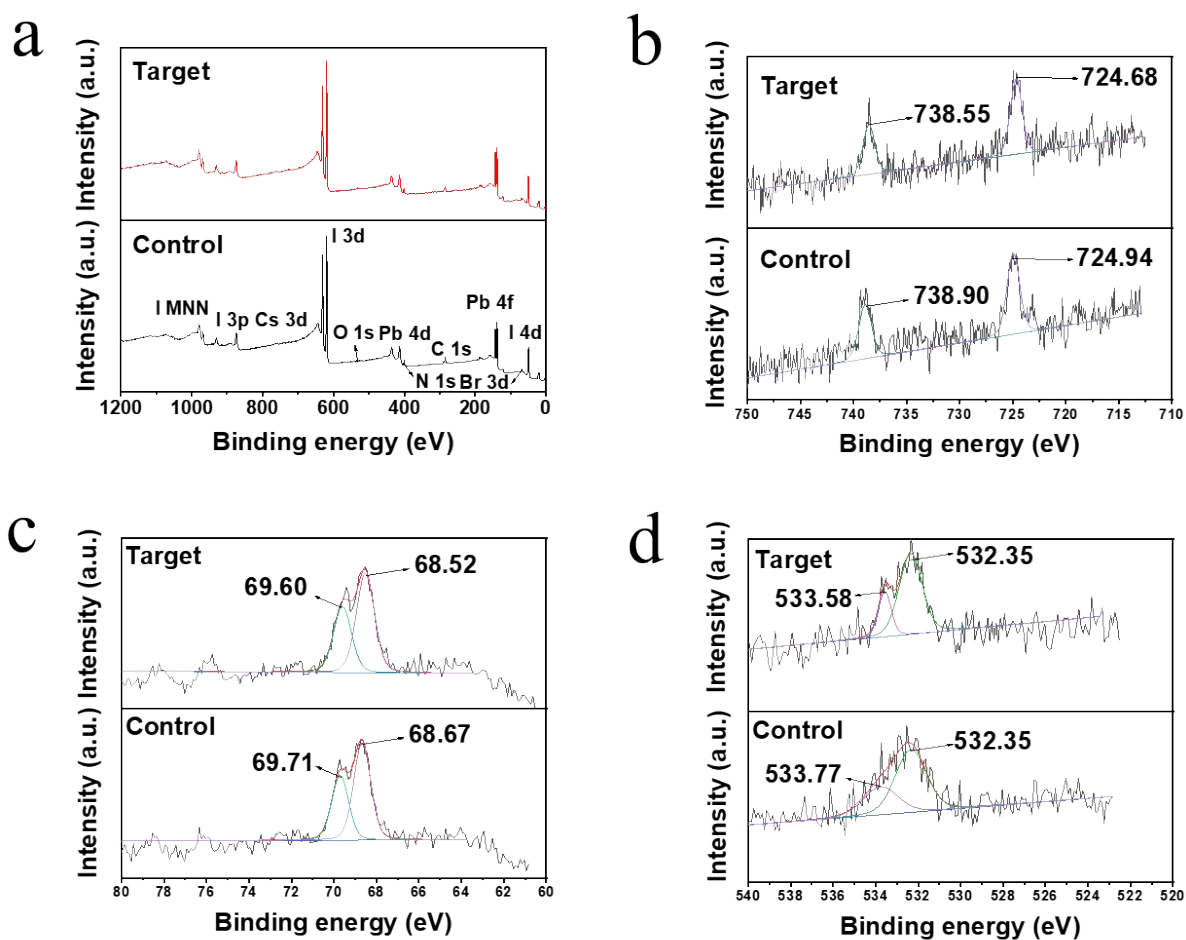
**Figure S1** J-V curves of different concentration



**Figure S2** (a) AFM images of the control perovskite film, (b) AFM images of the target perovskite film.



**Figure S3** Cross-sectional view FESEM images of PSCs assembled with (a) the control perovskite film and (b) the target perovskite film.



**Figure S4** (a) Survey spectra, (b) high resolution XPS spectra of Cs 3d, (c) high resolution XPS spectra of Br 3d, (d) high resolution XPS spectra of O 1s.

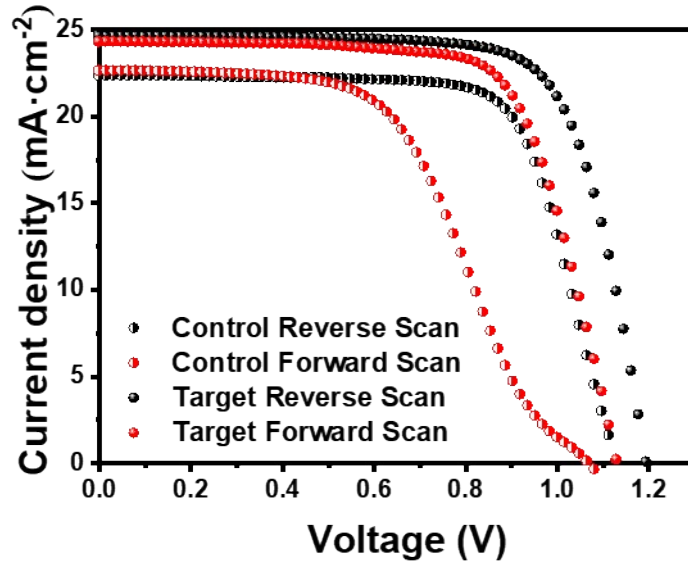


Figure S5 J-V curves in the forward and reverse scan of a random device.

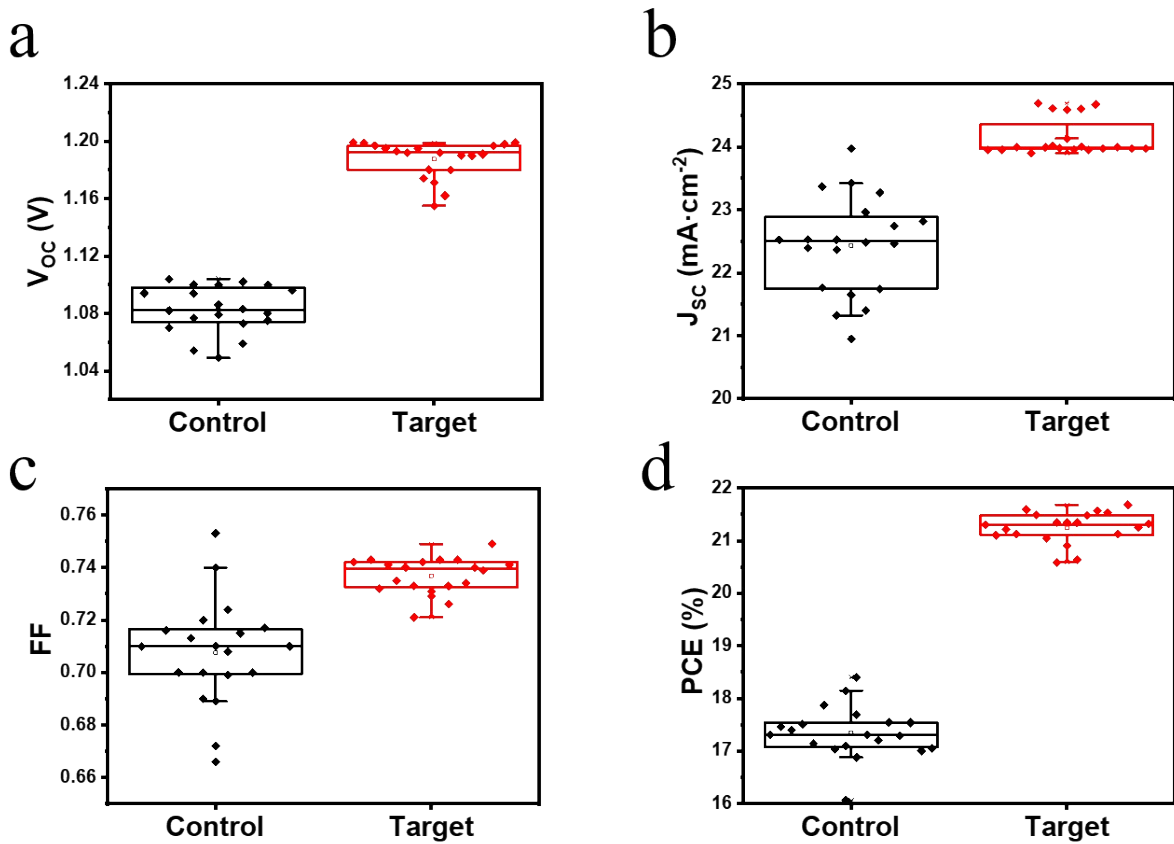
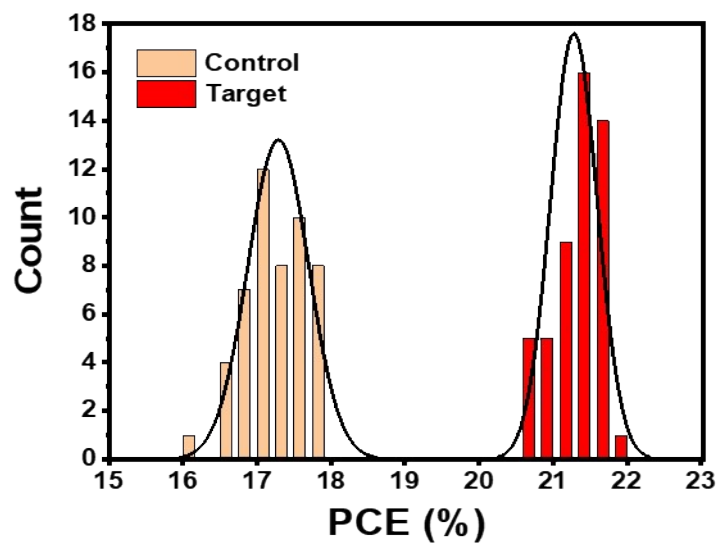
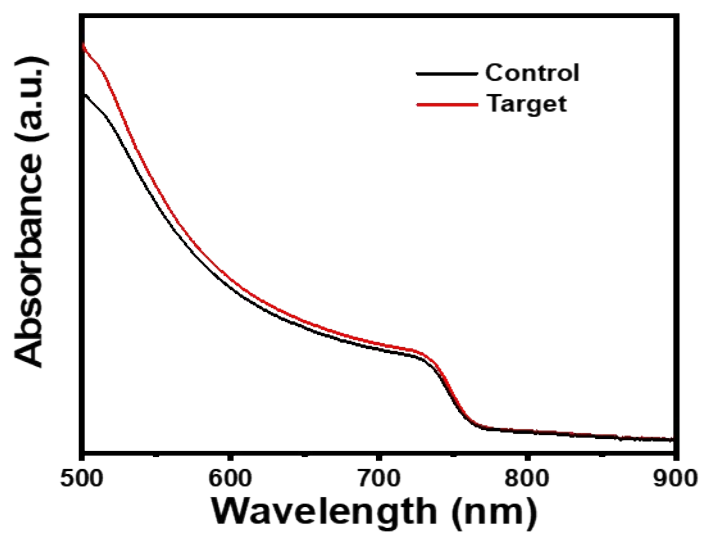


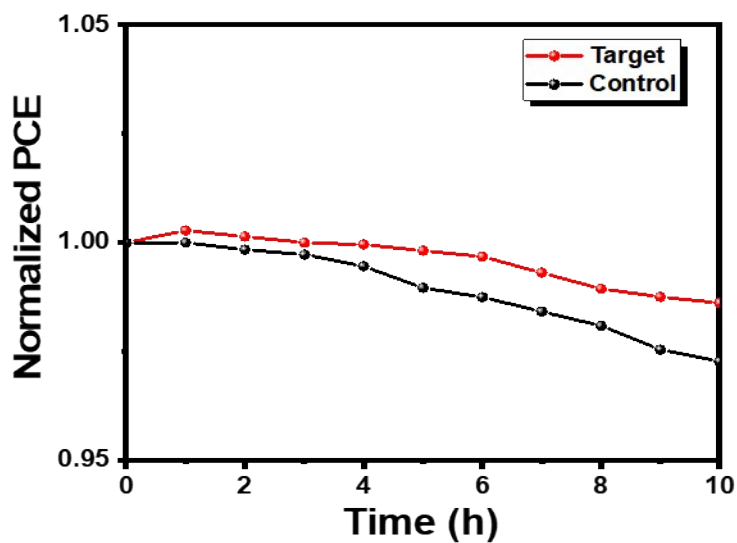
Figure S6 Box plots of the photovoltaic parameters for 50 independent PSCs fabricated under the same experimental conditions.



**Figure S7** PCE distribution of the control and target PSC devices.



**Figure S8** UV-vis absorption spectra for the control and target perovskite films.



**Figure S9** Light stability tests of the devices without encapsulation under AM 1.5G illumination.

**Table S1** The performance parameters of a random device in the forward and reverse scan.

Device	Scan Direction	$V_{OC}$ (V)	$J_{SC}$ ( $\text{mA}\cdot\text{cm}^{-2}$ )	FF	PCE (%)
<b>Control</b>	Reverse Scan	1.129	22.39	0.717	18.15
	Forward Scan	1.055	22.65	0.536	12.80
<b>Target</b>	Reverse Scan	1.209	23.95	0.749	21.68
	Forward Scan	1.129	24.32	0.707	19.42